

Delayed four- and six-wave mixing in a coherently prepared atomic ensemble

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We report on the simultaneous observation, by delayed Bragg diffraction, of four- and six-wave mixing processes in a coherently prepared atomic ensemble consisting of cold cesium atoms. For each diffracted order, we observe different temporal pulse shapes and dependencies with the intensities of the exciting fields, evidencing the different mechanisms involved in each process. The various observations are well described by a simplified analytical theory, which considers the atomic system as an ensemble of three-level atoms in Λ configuration. © 2010 Optical Society of America

OCIS codes: 270.1670, 190.4223, 020.1670

The transfer of light coherence to atomic quantum-state coherence is at the heart of many intriguing effects associated with light-matter interactions and has played a fundamental role for the enormous developments achieved in this field [1]. Coherently prepared atomic systems giving rise to electromagnetic induced transparency (EIT), coherent population trapping, slow and stopped light, have attracted much attention in recent years [2–4], specially owing to the possibility of storing and manipulating classical and quantum information in long-lived atomic coherences [1]. Nonlinear cw four-wave mixing (FWM) processes in EIT media have been extensively investigated [5, 6] and, in particular, they have been applied for the generation, in atomic ensembles, of both narrow-band photon pairs [7] and pairs of intense light beams showing a high degree of intensity squeezing [8]. On the other hand, multi-wave mixing processes have also been investigated using spatially resolved techniques [9, 10] and, more recently, the interference between cw FWM and six wave mixing (SWM) signals was demonstrated [11].

In this Letter, we report the observation of higher order nonlinear wave mixing processes in a coherently prepared cold atomic ensemble by means of the simultaneous emission of two pulses generated by delayed FWM and SWM processes. The delayed FWM pulse generation

is presently a well known phenomenon, which has been explored particularly for applications in all-optical routing [12] and in quantum information [13]. The delayed SWM pulse generation, on the other hand, was just recently predicted in [14] and here we provide its first observation, highlighting its difference with the corresponding FWM process by detecting both signals simultaneously. Different pulse shapes and dependencies on the incident field intensities are observed. All these features are predicted by a simplified theoretical model which allowed us to characterize the two pulses as associated with distinct nonparametric, nonlinear processes. The possibility of observing higher order nonlinear interaction between light and coherent atomic systems could play an important role for applications in quantum information, since they present quite different photon correlations. One goal, for example, is to generate quantum-correlated pulse pairs from such coherently prepared atomic ensembles [14]. The delayed FWM pulse pairs should exhibit correlations only in the saturated regime [14], the pulses being independent from each other at low powers. On the other hand, a pair of delayed SMW pulses should exhibit stronger correlations for any readout power, since they rely on all fields to be generated.

The theoretical analysis of the problem was developed in [14] and here we just review its main steps. We consider three-level atoms consisting of two degenerate Zeeman ground states and one excited Zeeman state interacting with two writing beams W and W' with a small angle between them and with opposite circular polarizations (Fig. 1). These two beams act for a long time so as to induce a stationary Zeeman ground state coherence grating before they are turned off. After the storage time t_s , the grating is read out by two reading beams R and R' , which also have opposite circular polarizations (Fig. 1). In our model, the stored coherence grating decays with a rate γ assumed to be much smaller than the excited state decay rate Γ .

As a result of the action of these four fields, optical coherences are excited in both transitions, resulting in the emission of two fields with opposite polarizations and propagating in the direction opposite to W' . In [14] we show that these optical coherences can be written as a sum of three terms, corresponding to the processes of stimulated emission, delayed FWM, and delayed SWM, respectively. Not all these processes, however, are phase matched and result in propagating optical fields. For the experimental situation considered here, note that the two reading beams are both counterpropagating with respect to W . This geometry for the readout fields is quite different from the one explored in [14], and results in the phase matching of one of the delayed SWM terms present in the locally excited optical coherences, together with one of the delayed FWM terms explored in [14]. Explicitly, from [14] we obtain that the terms of the local optical coherences contributing to these phase-matched, delayed

FWM and SWM signals are given respectively by

$$\sigma_{M_F-1, M_F}(t) \propto \frac{|\Omega_R|e^{-\gamma t_s}}{I_t \Gamma} [f_r(t)I_R + g_r(t)I_{R'}] \times e^{-i(\vec{k}_R + \vec{k}_W - \vec{k}_{W'}) \cdot \vec{r}}, \quad (1a)$$

$$\sigma_{M_F+1, M_F}(t) \propto \frac{I_R |\Omega_{R'}|e^{-\gamma t_s}}{I_t \Gamma} [f_r(t) - g_r(t)] \times e^{-i(2\vec{k}_R - \vec{k}_{R'} + \vec{k}_W - \vec{k}_{W'}) \cdot \vec{r}}, \quad (1b)$$

where \vec{k}_X (with $X = R, R', W$, or W') are wavevectors associated with the reading and writing beams, considered as plane waves all having the same frequency, resonant with the atomic transition. These expressions are valid for γ much smaller than the Rabi frequencies Ω_X [14]. Since $\gamma \ll \Gamma$ for our experimental conditions, they describe both low-intensity and saturated regimes of these processes. For simplicity, we omitted the dependence with $\Omega_W, \Omega_{W'}$ from these expressions. An analysis of their low-intensity limit reveals the usual dependences of $|\Omega_W||\Omega_{W'}||\Omega_R|$ and $|\Omega_W||\Omega_{W'}||\Omega_R|^2|\Omega_{R'}|$ for the FWM and SWM processes, respectively. In Eqs. (1), I_R ($I_{R'}$) corresponds to the intensity of beam R (R'), and $I_t = I_R + I_{R'}$. Functions $f_r(t)$ and $g_r(t)$ are defined in [14]. They depend only on the parameters I_t and Γ and determine the temporal pulse shapes associated with each term of the optical coherences.

The delayed FWM signal in the above equations can be interpreted as originating from Bragg diffraction of beam R from the ground state coherence grating created by the writing beams. On the other hand, the SWM signal is a higher order process originated from Bragg diffraction of beam R from population gratings created in the ground and excited states of the corresponding transition, by the simultaneous action of both R and R' over the original coherence grating induced by the writing beams W and W' . These population gratings responsible by the SWM signal require then all four incident fields in Fig. 1(a) to be created, while the coherence grating responsible by the FWM signal requires only fields W, W' .

Since we consider a system composed of Zeeman sublevels, each transition corresponds to a well defined selection rule. Therefore the beams generated in each transition have orthogonal circular polarizations, an essential aspect for separating each signal experimentally. Neglecting propagation effects, the electric field and the intensity in each mode can be promptly calculated [15]. Experimentally we focus attention on the pulse shapes associated with the FWM and SWM processes (proportional to the squared modulus of Eqs. (1a) and (1b), respectively) and on the corresponding retrieved energies. The expressions for the retrieved energies in each pulse, that will be used to compare with our experimental results, can be

readily obtained [14, 15] from Eqs. (1a) and (1b):

$$U_{FWM} \propto \frac{I_R}{I_t 2} \int_0^\infty |f_r(t)I_R + g_r(t)I_{R'}|^2 dt, \quad (2a)$$

$$U_{SWM} \propto \frac{I_{R'} I_R^2}{I_t 2} \int_0^\infty |f_r(t) - g_r(t)|^2 dt. \quad (2b)$$

The experiment is performed using cold cesium atoms provided by a magneto-optical trap (MOT). The atoms are initially pumped into the $6S_{1/2}(F = 3)$ ground state using the method described in [15]. The MOT temperature was estimated in the range of mK and the atomic density in the lower ground state is $n \approx 10^{10} \text{ cm}^{-3}$. The MOT quadrupole magnetic field is switched off while three pairs of Helmholtz coils are used in order to compensate for stray magnetic fields. For the writing and reading beams we employ an external cavity diode laser locked on the cesium closed hyperfine transition $6S_{1/2}(F = 3) \rightarrow 6P_{3/2}(F' = 2)$, with the time sequence determined by acousto-optical modulators. The opposite circular polarization states for the pairs of writing and reading beams are obtained by using polarizing beam splitters (PBS) and quarter waveplates. The grating storage time t_s is fixed and equal to $1 \mu\text{s}$. The manipulation and storage of such gratings in our system, as a function of t_s , were investigated in more detail in [15, 16]. The retrieved signals associated with the delayed FWM and SWM can be separated with a quarter waveplate followed by a PBS as shown in Fig. 1(b).

In Fig. 2(a) we show the temporal shape of the retrieved pulses associated with each wave mixing process. The corresponding powers for the writing (W , W') and reading (R , R') beams are approximately equal to (20 mW/cm², 2 mW/cm²) and (10 mW/cm², 10 mW/cm²), respectively. Clearly, the different observed pulse shapes strongly suggest the two signals originate from different mechanisms. In Fig. 2(b) we plot the theoretical pulse shapes associated with each retrieved signal. In order to compare directly with the experimental results, we convoluted the pulse shapes provided by Eqs. (1a) and (1b) with the time response of our detectors (PDA36A from Thorlabs), described by a single response-time parameter τ [17]. From such convolution we estimate $\tau = 0.2 \mu\text{s}$, consistent with the detector's bandwidth.

In another series of experiments, we fixed the total intensity I_t associated with the reading beams and changed the relative values of I_R and $I_{R'}$ (so that $I_{R'} = I_t - I_R$). In Fig. 3 we show the retrieved pulse energy associated with the FWM and SWM processes as a function of the intensity of field R normalized by I_t . As expected, while the extracted energy associated with the FWM process monotonically increases with the reading beam intensity I_R , the similar quantity associated with the SWM process vanishes at the extreme values of the intensities corresponding to ($I_R = 0$, $I_{R'} = I_t$) and ($I_R = I_t$, $I_{R'} = 0$), respectively. This behavior is consistent with the predictions of Eq. 2(b), which require all fields to be present

for the SWM signal to be observed. The theoretical curves, solid lines, describe quite well these observations, together with the relative values of the retrieved energy for each process. Even though, they fail to describe the observed symmetry of the curve related to U_{SWM} . We attribute this discrepancy to the simplicity of our model with respect to the real Zemann structure in the experiment.

In summary, we have demonstrated experimentally the observation of delayed four- and six-wave mixing in a coherently prepared cold cesium ensemble. The measured results are in qualitative agreement with the predictions of a simple theoretical model for an ensemble of three-level atoms. Our results clearly demonstrate the possibility for distributing previously stored light information in different optical modes and through different nonlinear processes, which might be of considerable interest for the growing field of quantum information.

This work was supported by the Brazilian agencies CNPq and FACEPE and the National Institute of Science and Technology for Quantum Information.

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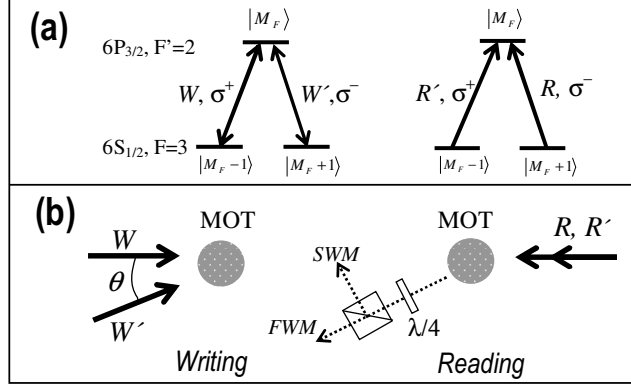


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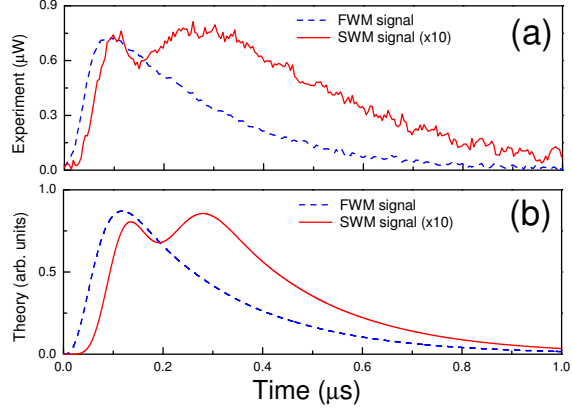


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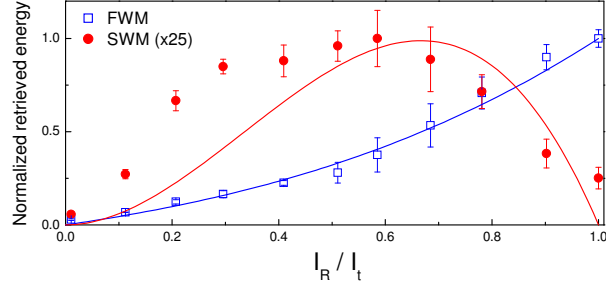


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